INFLUENCE OF THE STRUCTURE DISORDER ON THE HOLE MOBILITY IN THE TETRACENE LAYERS.

S. Kania, J. Kondrasiuk, M. Wieczorek*

Institute of Physics, Technical University of Łódź, Wólczańska 219, 93 – 005, Łódź Poland.
*Institute of Chemistry and Environmental Protection, Pedagogical University,
Armii Krajowej 13/15 av., 42-201, Częstochowa, Poland

The hole drift mobility measurements in the quasi - amorphous tetracene films were carried out, and for comparative purposes it was made revision for the hole investigations in the polycrystalline tetracene films. The quasi – amorphous tetracene layers were obtained with aid of vacuum evaporation technique. Tetracene was deposited on the glass substrates with the temperature 170 - 200 K in vacuum of the order of 10^{-5} Torr. Whereas polycrystalline tetracene layers were obtained with use of the evaporation technique as well. Here, the tetracene was also deposited on the glass substrate(in the room temperature) in the vacuum of the order of 10^{-5} Torr. The holes mobility was evaluated with the aid of the time – of – flight method. Obtained results suggest that we are with hopping transport throughout localized states near the Fermi level. Furthermore, in the slight manner, the studies demonstrate a little influence of the structure order on the holes mobility value, whereas it was not state the influence of the structure on the transport character for the holes in the tetracene films.

INTRODUCTION

The fundamental parameter characterized mechanism of the transport in organic and inorganic solid is drift carrier mobility. That is why this characteristic feature is studied under so great intensity in so many centers.
        Charge transport in organic molecular materials differs significantly from that of inorganic substances. This is due to the comparatively weak short-range intermolecular forces which enable a single molecule to preserve its individuality to some extend. The mean free path of the electron in anthracene is estimated to be of the same order as the intermolecular spacing in anthracene, so it might be not realistic to describe charge carrier motion in terms of the band model. Taking into account the temperature dependence of both hole and electron drift mobility in single crystals of simple aromatic hydrocarbons, one may suggest, that in general the band model describes well the charge transport in these materials, though in the case of naphtalene single crystals a remarkable change of the temperature dependence of electron mobility at about 80 - 100 K has been detected [1]. It has been suggested that it may be a result of band – to – hopping transition, i.e. the band transport has been suggested to dominate at the lower temperatures and the hopping transport has been supposed to dominate at

the temperatures higher than about 150 K. However, in the case of charge transport in polycrystalline structures of simple aromatic hydrocarbons (and as tetracene, coronene, p-terphenyl, and p-quaterphenyl), hopping transport among localized states proved to be dominant [2-4]. In this paper the results of measurements of holes drift mobility in quasi - amorphous tetracene layers are presented.

EXPERIMENT

The tetracene quasi – amorphous samples were obtained by evaporation in vacuum under the pressure of the order of 10^{-5} Torr on glass plates covered with metal film. The substrate temperature was from 170 to 200 K and the evaporation rate was changed in the range 80 – 110 Å/s. Structural examination of the obtained tetracene layers was made using X – ray diffraction. In order to attain that, one employed an automatic diffractometer DAR. Diffraction examinations were made in the 2θ range from 5° to 80° with measuring step 0,05°.
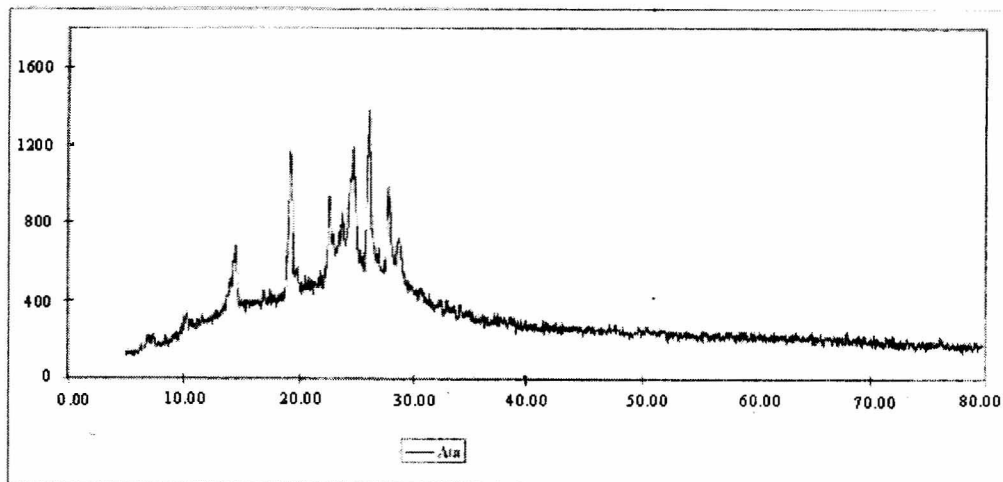Typical diffraction pattern for the quasi - amorphous tetracene films is described with Fig. 1.

Figure 1. Diffraction pattern (X – ray) for quasi – amorphous tetracene film. Plot of the intensity of diffraction lines in function of the angle 2Θ.

There is a clear evidence in this diffraction graph vote for partial appearance of polycrystalline phase and for partial existence of long range order.

Whereas the fig. 2. describes typical diffraction pattern for polycrystalline tetracene structure. The polycrystalline tetracene layers were obtained by evaporation in vacuum under the pressure of the order of 10^{-5} Torr on glass plates covered with gold film. The substrate temperature was at about 300 K and the evaporation rate was changed in the range 20 – 30 Å/s.

Structural examination of the obtained polycrystalline films was made as identically as for quasi – amorphous layers.
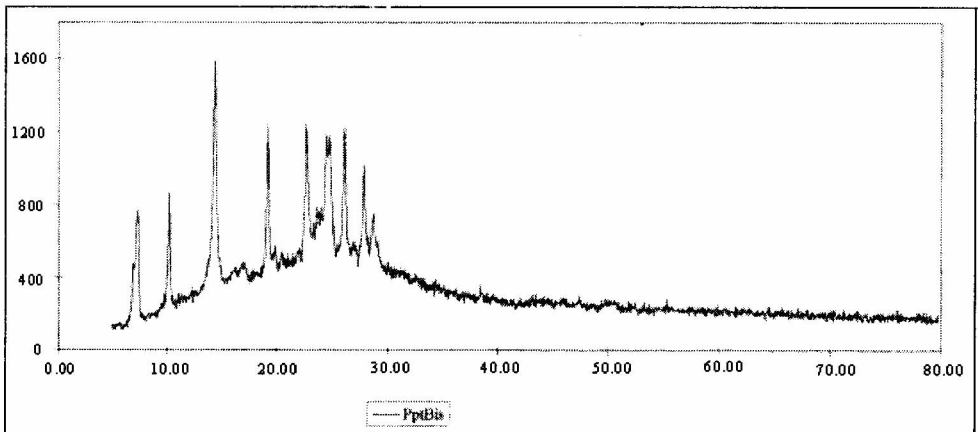

Figure 2. Diffraction pattern (X – ray) for polycrystalline tetracene film. Plot of the intensity of diffraction lines in function of the angle 2Θ.
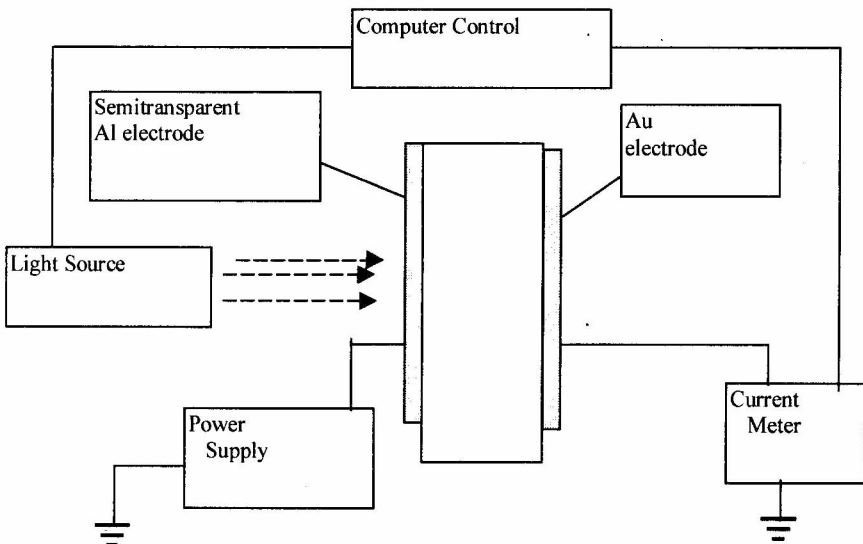

Figure 3. Schematic diagram of the equipment used for the time–of–flight measurements

The Fig. 1. makes one clear that the long – range order is visible, i.e. the obtained structure is similar to the structure of the amorphous carbon films [5].

In order to attain chance to make the investigations of the electrical properties for obtained tetracene layers, the semitransparent Al electrodes were also evaporated in vacuum.

Fig. 3 shows a schematic diagram of the equipment used for the time – of – flight method measurements. A short light pulse goes through the semitransparent aluminum electrode and generates a number of electron – hole pairs in a thin layer at the electrode.

Depending on the polarization of sample, either electrons or holes travel through a sample giving rise to a current signal registered by a current meter.

All the system works under computer control which enables to control the measurements and to store the data. The time of flight is found from the current signal. The current pulses were measured with digital oscilloscope DSO 5804.

RESULTS AND DISCUSSION

Typical current pulses for hole obtained for the quasi - amorphous tetracene films are shown in the Fig. 4, whereas the current pulses for hole in polycrystalline tetracene are shown in the Fig. 5.
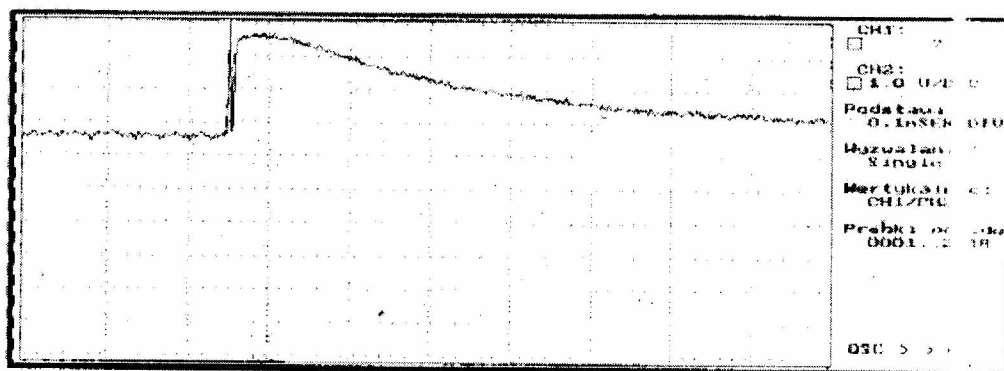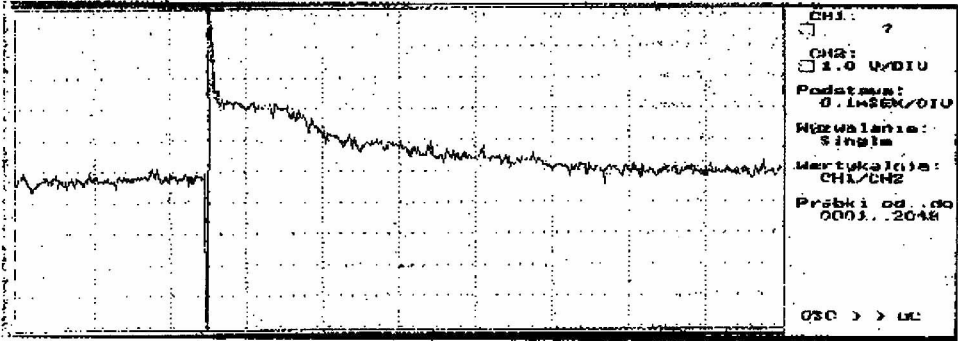

Figure 4. Typical current pulse for holes obtained for the quasi – amorphous tetracene film, thickness L = 17,5 μm, voltage applied to the film U = 14 V.

Figure 5. Typical current pulse for holes obtained for the polycrystalline tetracene film, thickness L = 20 μm, voltage applied to the film U = 7 V.

The holes drift mobility for quasi – amorphous and for polycrystalline layer is calculated from the expression:

$$\mu = L^2/t_r \cdot U, \tag{1}$$

where: L is the sample thickness, t_r is time of flight and U is the voltage. In order to attain chance to evaluate the faultiness of the estimation of the time-of-flight, one performed a diagram of an inverse time-of-flight $1/t_r$ upon the voltage applied to the film. The plots of those dependences are presented in the Fig. 6 for quasi – amorphous layers.
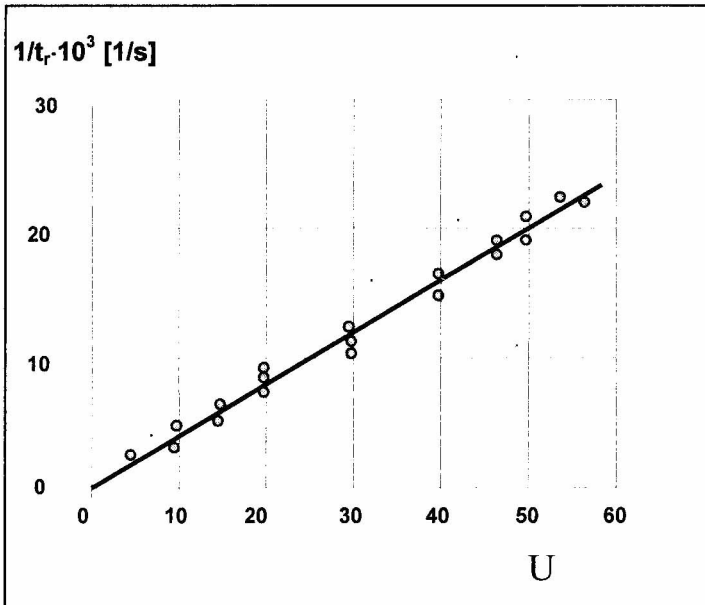

Figure 6. Diagram of an inverse time-of-flight $1/t_r$ for holes (in quasi – amorphous layer) upon the voltage applied to the film,   L = 17.5 μm, μ_h = $1.2 \cdot 10^{-3}$ cm^2/Vs.

The voltage dependence of the time – of – flight $1/t_r$ for the polycrystalline layer is presented in the Fig. 7. Obtained current pulses had characteristic kink point which enables direct estimation of the transit time for holes both for polycrystalline layers and for quasi – amorphous layers.
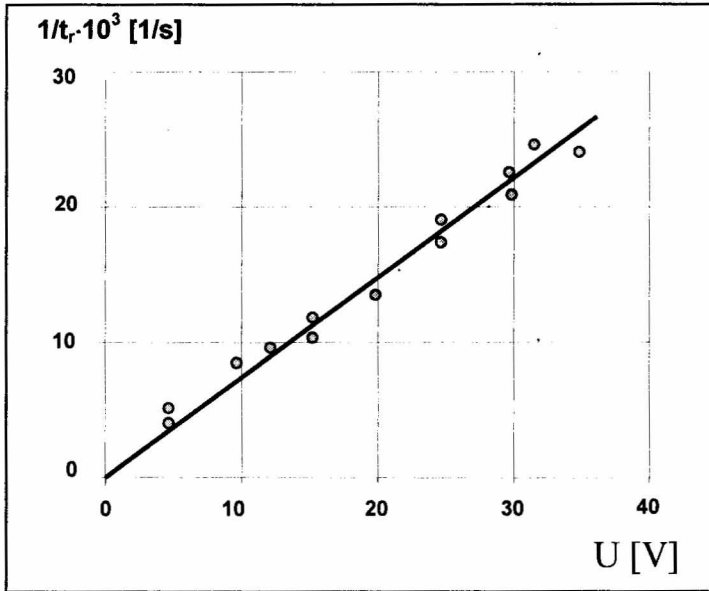

Figure 7. Diagram of an inverse time-of-flight $1/t_r$ for holes (in polycrystalline tetracen layers) upon the voltage applied to the film,   L = 18,5 μm, $\mu_h = 2,59 \cdot 10^{-3} cm^2/Vs$.

The temperature dependence of the drift mobility for holes was measured in the temperature interval 295 ÷ 345 K, and the activation energy for mobility was determined to be 0,025 - 0,03 eV. Similar activation energy dependence, in the interval width of the kT, was obtained for the polycrystalline layers.

The activation energy of the drift mobility was of the order kT and low drift mobility for holes in quasi – amorphous and polycrystalline tetracene layers suggests that the mode of hopping transport in narrow band of localized states and on Fermi level should be taken into account.

Wide spread of holes mobility values for quasi - amorphous tetracene layers (the number of the layers was fifteen) was observed and the mobility value for them was in the range from $5,4 \cdot 10^{-4} – 2,6 \cdot 10^{-3}$ cm^2/Vs. For polycrystalline tetracene layers the mobility was $(2,59 \pm 0,45) \cdot 10^{-3}$ cm^2/Vs. The investigations of the hole mobility have been made in the air with the humidity of the order of 70 %. On the basis of the obtained results one can suggest, that rise of the structure disorder has the influence in the mobility range, but no more then circa one half of the order of magnitude .
Relatively great rise of the mobility value in the case of the quasi –

amorphous tetracene films could be due to the thermal no stability of the films.

Whereas, there was not stated the influence of the structure disorder on the current impulse shape. For both of the cases the quasi – Gaussian shape of the carrier packet was observed. There was not observed the transformation from the Gaussian transport into dispersive transport, what one can expect for quasi – amorphous layers. The shape of the current pulse, which is presented in the Fig. 4 and 5, testify in favor of it.

The drift mobility in this model is given by [7]:

$$\mu = (1/6)\, \nu_{ph}\ (\ R^2 e/kT)\cdot\ \cdot\exp(-2\alpha R)\cdot\exp(-\Delta E/kT), \qquad (2)$$

where: ν_{ph} – is the phonon frequency, R – is the average distance between localized states, e – is electron charge, , α - is the decay of the localized state wave function, ΔE – is the width of the narrow band of localized states due to the molecule structure which takes part in the charge transport. Usually α^{-1} may be assumed to be of the order of the average distance between molecules. The choice of α^{-1} is burdened with some uncertainty. Using the expression (2) for the carrier mobility and geometric formula for density of states:

$$N(E_F) = (3/4\pi)(1/R^3\Delta E) \qquad (3)$$

one can estimate the average distance between the localized states and the density of localized states at the Fermi level. As it was already mentioned, the spread value in the hole mobility, for the quasi – amorphous tetracene films, would have been make with the structural changes during performing observations. There is the reason of our opinion that these studies must be continued. Based on the formula (2) and (3) and took into consideration obtained mobility data for hole in the quasi – amorphous and polycrystalline layers it was made an estimation of the average distance between localized states R_h and the localized states concentration at the Fermi level.

CONCLUSIONS

There is a possibility to formulate some conclusions on the basis of our investigations for the hole mobility in the quasi – amorphous   and polycrystalline films:

obtained hole mobility in polycrystalline layers was $(2,59 \pm 0,45)\cdot10^{-3}$ cm^2/V-s;

obtained hole mobilities in quasi – amorphous layers were from $5,4\cdot10^{-4}$ to $2,6\cdot10^{-3}$ cm^2/Vs;

activation energy for mobility for quasi – amorphous and polycrystalline layers is in the order of kT.

Taking mentioned above into account, we can fall in the suggestion that the dominant transport mechanism, here, may be hopping transport.

Beyond that it appears, that:
the magnitude of the disorder has a certain influence on the hole mobility value in the tetracene layers;
there was no observed <u>distinguishable</u> influence of the disorder on character of the transport with the packet of the carriers, i.e. the dispersive transport of the carrier packet has not been observed.

ACKNOWLEDGEMENTS

REFERENCES

[1] E.A. Silinsh, Organic Molecular Crystals, Springer – Verlag 1980.p.37.
[2] S.Kania, W. Mycielski, A. Lipiński "Charge carrier transport in oriented p-quaterphenyl layers", Thin Solid Films, 61, 229 (1979).
[3] A. Lipiński, W. Mycielski, J. Świątek, "Charge carrier transport and D.C. conductivity in thin polycrystalline p – terphenyl films", J. Chem. Solids 41, 455 (1980).
[4] W. Mycielski, Evidence of hopping conduction among localised states in disordered systems of simple aromatic compounds", J. Non – Crystalline Solids 37, 267 (1980).
[5] S. Kania, J. Kondrasiuk, "charge carrier mobilities in the tetracene layers in presence of the oxygen molecules (O_2) and water vapour (H_2O)", ,Scientific Bull. of Technical Univ. of Łódź 16, 43 (1996)
[6] R. Zahlen The Physics of Amorphous Solids, 1983, John Wiley and Sons Inc.
[7] N. F. Mott and E. A. Davies, Electronic Processes in Non – Crystalline Materials, Clarendon press, Oxford (1971).
[8] S. Kania, J. Kondrasiuk, G. W. Bąk, "Influence of ambient atmosphere on charge transport in thin films of low – molecular weight organic compounds", Molecular Phys. Reports, 25, 93, (1999).